Annual Work Proposal
Office of International Health Programs (EH-63)
U.S. Department of Energy

Project Indalo

Centro de Investigaciones Energeticas Medioambientales y Tecnologicas (CIEMAT)

Purpose: In 2002, we will continue to perform the radiological medical surveillance and environmental monitoring in the area of Palomares (Almería - Spain). In addition it is intended to continue activities included in the recommendations made by the expert panel in its 1998 report.

As reported in the 2001 Annual Work Report, a change in circumstances requires a change in the management of the Palomares situation. The new management plan, that is being negotiated, involves a process of ownership conveyance (from the current owners to an administrative body) of the lands where the higher residual contamination levels from the 1966 accident remain. This process is essential in order to assure the implementation of further research activities in the more affected zones. The most suitable way to achieve the ownership conveyance is being discussed among representatives of the national regulatory body Consejo de Seguridad Nuclear, CIEMAT, ENRESA (National Agency for the Management of Radioactive Wastes) and the municipality of Palomares. Then, a research plan will be carried out. In addition to scientific objectives, leading to improve the knowledge on environmental and metabolic behavior of Pu and Am, the final goal of the research plan is to determine the most suitable options for environmental restoration. Obviously, this should be based on risk assessments under present conditions and other potential future scenarios of use.

During 2002, the preliminary phase of the research plan, mentioned earlier, should be started. This phase mainly consists in updating and improving the radiological characterization of the global area of Palomares. The recent finding of several significant contaminated metallic fragments with adhered soil particles in a part of the zone 3, close to the impact point of the bomb 3, but in the opposite direction to the original contamination plume is one of the reasons to do it. The results of this phase should provide reliable information leading us to take a confident decision regarding purchase of land pieces *(extension and location)*, other than zone 2-0. The planned radiological characterization will also contribute to the identification of hot points of secondary accumulation of the contamination through wind or water erosion. Work in this phase will combine extensive radiometric surveys and soil sampling and analyses.

In relation to air monitoring, sampling and analyses will continue during 2002 at three of the established locations (stations 2-1, 2-2 and P). Work in station 2-0 will depend on two conditions, availability of the new air pump and permission of the owner of the plot where the station is located.

Concerning vegetation studies, it is intended to start the historical review data analysis, postponed in 2001 due to other work priorities, in order to refine risk assessment for the ingestion pathway. In addition, the outstanding vegetable samples collected during 2000

and 2001 will be analyzed. During 2002 a sampling of vegetables in the most potentially affected zones will be performed.

Progress in the statistical significance of Pu and Am content in relevant animal consumption products will continue by sampling and analyses of cow and goat milk and snails. The remaining analyses of snails will be also performed.

Effort on hot particles will continue to put emphasis in their separation from soil samples and in the determination of their realistic isotopic composition. Also activity estimates and their contribution to the soil activity will be determined.

Risk assessment will be an important issue of the research program mentioned earlier, which is presently being designed as a part of the new management plan in Palomares. Therefore, this study will be probably resumed in 2003.

The following is a general description of work to be performed.

General Description of Work to be Performed:

I. Concise Statement of Goals

The former goals remain unchanged since the signing of the Hall-Otero Agreement. These are:

- To determine the magnitude of the risk of internal contamination in the inhabitants of the zone during the period immediately following the accident and the subsequent emergency phase.

- To assess the short, medium, and long-term risk of internal contamination for those people living in and around Palomares, those who cultivate the contaminated land and those who consume vegetable products grown in this area, as well as products from animals which have been given cereals and other vegetables grown in the area as fodder.

In addition to these objectives and as a consequence of the new management plan that is being negotiated, it is intended to implement a research plan with the final goal of determining the most suitable options for environmental restoration.

II. Background (includes relevance to DOE programs)

As a consequence of the accident which occurred on January 17, 1966, a radiological medical surveillance and environmental monitoring program has been conducted in Palomares, Spain. This work has been performed pursuant to the Hall-Otero Agreement of February 25, 1966. In Spain, the Centro de Investigaciones Energeticas Medioambientales y Tecnologicas (CIEMAT; Center for Energy, Environmental, and Technological Investigations) is the organization responsible for all technical aspects of this project. CIEMAT provides semi-annual reports to the Spanish Consejo de Seguridad Nuclear (CSN; Nuclear Safety Board), which, in turn, provides summaries of the activities in Palomares to the Spanish Parliament. The CSN is the Spanish organization responsible for radiation protection in general.

III. Methods and Approach

The medical monitoring program consists of clinical examinations and radiobioassays of Pu and Am collected from 24-hour urine samples of 150 residents from Palomares every year. The individuals examined differ each year unless some Pu or Am in urine was detected from the previous year's examination. If necessary, individuals with high potential internal contamination are examined by CIEMAT´s whole body counter. The details of the clinical examinations are on record at DOE. The examinations are performed during the spring and autumn of each year. Approximately 10 individuals are examined each week.

The environmental monitoring program consists of sampling, analysis, and measurements of Pu and Am in air, soil, food crops, wild vegetation, milk, and other products. For example, the air is sampled continuously by high volume samplers equipped with a PM-10 inlet. The filters are changed weekly. There are four air high volume continuous samplers in the area. Soils are sampled with a frequency depending on the characteristics of the experiment to be performed. For example, the frequency of deep soil samples is less than surface soil samples. The frequency of samples in vegetables depends on the growing season and the vegetable species cultivated each year.

Air and urine samples are analyzed by sequential radiochemistry methods for Pu and Am. On separate planchets, Pu and Am are then measured by alpha spectrometry. Other samples, such as soils and vegetation, are analyzed for Pu by radiochemistry and then measured by alpha spectrometry. However, Am is measured directly (with previous drying and removal of organic matter for soils) by gamma spectrometry without previous radiochemistry. Need to improve the detection limit of Am activity for vegetable samples, changed the direct gamma spectrometry measurements by radiochemistry and alpha spectrometry at least for the most representative samples. For milk samples, analyses of Pu and Am by sequential radiochemistry methods are done and then alpha spectrometry measurements are decided. All radioanalyses and measurements are performed according to established procedures at CIEMAT.

The specific sampling, analysis and assessment plan for 2002 is presented below:

Soils:

As mentioned above, work on soils during 2002 will combine extensive radiometric surveys and soil sampling and analyses, dealing with the updating and improvement of the radiological characterization of the global area of Palomares.

<u>Radiometric survey</u>
Specifically, during 2002 radiometric surveys will be carried out in the following zones:

- Zone 3: The survey carried out during 2001 will be extended to cover other adjacent plots to the impact point of bomb 3. Even a new survey in the 2001 already controlled surface would be necessary if a new ploughing takes place.

- Influence of run-off from zone 2-0: Some recent data coming from the study of the influence of run-off in zone 2-0, suggest that the contamination from this zone is being dispersed beyond the originally considered zero line of contamination. The magnitude and extension of this contamination will be investigated by a radiometric survey of the zone.

Soils sampling and analyses

Presently it is clear that the activity in the soils of Palomares is not homogeneously distributed. Depending of its location, soils contain different amounts of hot particles, fine dispersed contamination and, even, metallic debris. The achievement of representative results when the activity is not homogeneously spread, as consequence of the presence of hot particles, is a problem to be solved because the soil sample size that is normally processed by radiochemical separation procedures is much lower than the related soil sample collected. Two approaches can be established to solve the problem of representatively for soils collected in areas with hot particles:

- To implement an analytical procedure able to process large amounts of sample, representative of the total sample collected. At present, there are no good methods describing total dissolution of large soil samples.

- To increase the number of soil sub-samples for analysis, from each collected sample. The way to obtain the suitable number of sub-samples and analysis for the adequate representative samples should be designed. This approach will involve a large amount of laboratory work due to the complexity of the previous radiochemical separation method required for the Pu alpha spectrometry measurement. An indirect estimate of Pu activity by directly measuring Am-241 (low energy gamma spectrometry) and then applying the isotopic ratio Pu/Am can be used to minimize chemical efforts. This approach requires a good assessment of such ratio. A statistical approach managing data from Am activity in the analyzed soil sub-samples has to be designed, as well.

Since the previous inventory assessments were based on a homogeneous distribution of activity in each analyzed soil sample, this new methodological approach will allow confirm and refine the existing inventory and its distribution.

During 2002, the new approach will be applied to the soils collected in 2001 in zone 3 (including part of the zone 5). The analyses of soils adhered to the metallic fragments, found in this zone, will be continued to progress in determining the isotopic composition of the bomb 3.

Distribution of the contamination in relation to granulometric fractions in soils

The distribution of the contamination in soils is expected to be dependent of the soil location, not only due to the particular soil composition but also related to the proximity to the impact points. This distribution is being studied (initial data has been reported in the 2001 Annual Report) and could help to interpret the low level of risk through the inhalation pathway, calculated using results from air samples, in relation to the activity measured in soils. Activity in soils is higher near the impact points, where contaminated particles should be also larger in size. Far away, even outside the zero line, were wind dispersion could be significant, the soil contamination is expected to be concentrated in finer soil fractions. The distribution of the contamination can be also time dependent if

some type of hot particles corrosion is assumed. This could contribute to enhance the risk of inhalation with time and, probably, also the bioavailability for the food pathway. During 2002, the study of granulometric composition will progress by obtaining the specific activity in the different soil fractions in zone 2-0, in a sample collected in 1999 at the same location that one collected and studied in 1986. Therefore, the results should illustrate the temporary evolution of the distribution of the contamination in relation to granulometric fractions in soils after 25 years.

Hot particles

Radiochemical analysis of the isolated hot particles from soil adhered to one of the fragments found in zone 3 (potentially belonging to bomb-3), will be carried out to determine its isotopic composition. Also the activity associated with these hot particles and the contribution to the soil activity will be estimated.

Air Samples:

Weekly samples from 3 stations will be collected (meaning about 150 samples during the year in an optimal way). Aditional samples from another station could be obtained if the new air pump is received during 2002 and the permission of the owner of the plot where the station is located is obtained. However, unexpected stops of the samplers and difficulties of repairing in situ, could lead to the collection of a lower number of samples. During 2002, the total of the samples corresponding to 2001 sampling will be analyzed for Pu (130 samples). Also, about 50 samples corresponding to 2001 sampling will be analyzed for Am by radiochemistry and measured by alpha spectrometry (about 50 samples). The samples are analyzed individually and accumulated in a monthly basis for measurement.

Vegetation Samples:

The historical review data analysis, postponed in 2001 due to other work priorities, will start during 2002. This should allow to refine risk assessment for the ingestion pathway. The collection of vegetation will have a low priority during 2002. However a limited number of representative samples will be collected in the most potentially affected zones as a part of the routine area surveillance program. Also, vegetable samples collected during 2000 and 2001 will be analyzed, meaning about 30 analyses and further measurements.

Urine samples:

In the same way as in preceding years, 150 people from Palomares will be transported to CIEMAT headquarters in Madrid for medical examinations, sampling of 24-hour urine collections and further bioassay analyses leading to internal dosimetric assessments. In total, 300 samples will be analyzed, 150 for Pu and the same 150 for Am by radiochemistry followed by alpha spectrometry.

Other:

About 8-10 milk samples (half from cows and half from goats) will be collected and analyzed by radiochemistry and alpha spectrometry for Pu and Am during 2002. The remaining analyses of snails collected in 2001 will be also performed. This work will contribute to improve the statistical significance of Pu and Am content in relevant animal consumption products.

IV. Milestones and Deliverables (include dates)

By December 31, 2002, we intend to achieve the following Milestones:

- Perform clinical examinations and radiobioassays of Pu and Am collected from 24-hour urine samples of 150 residents from Palomares.

- Perform sampling, analysis, and measurements of Pu and Am in air, soil, food crops, wild vegetation, milk, and other products. The number of analyses is listed above in Section III.

- Perform specific activity in the different soil fractions in zone 2-0.

- Perform separation, identification, characterization and isotopic composition of hot particles in soils.

V. Suggested Performance Indicators

- Provide the individual results from clinical examinations and radiobioassays of Pu and Am collected from 24-hour urine samples to the 150 residents from Palomares who were examined during the year, by April 2003.

- Provide the results from the sampling, analysis, and measurements of Pu and Am in air, soil, food crops, wild vegetation, milk, and other samples to CSN by September 2002 (semi-annual report for the first half of 2002) and by March 2003 (semi-annual report for the second half of 2002). These reports will also be provided to DOE. The number of analysis is listed above in Section III.

- Provide the temporary evolution of the distribution of the contamination in relation to granulometric fractions in soils of the zone 2-0 after 25 years.

- Provide results concerning hot particles in soils to CSN and DOE by April 2003.

VI. References

Radiochemical analytical procedures were provided to DOE in 1992. Please see Annex I, Methodologies for Analysis and Measurements in "Summary Report on the Palomares Surveillance Program," July, 1992.

VII. CV's of Investigators (short 1-2 pages, if possible, including only relevant publications for the last 5-10 years)

Short CV's of José Gutiérrez, Asunción Espinosa, Antonio Aragón and Javier Martínez were submitted in 1999.

VIII. Budget Request (see attached form)

IX. Addendum Containing Relevant Publication Preprints, etc

- Vigilancia Radiológica en la Zona de Palomares. Informe al Consejo de Seguridad Nuclear (Segundo Semestre del Año 2001) CIEMAT/DIAE/PPRI/51100/02-2002.

- Vigilancia Radiológica en la Zona de Palomares. Informe al Consejo de Seguridad Nuclear (Primer Semestre del Año 2002)

- Tesis doctoral: Comportamiento ambiental de las partículas del combustible nuclear (fundamentalmente plutonio) tras un accidente nuclear en un ecosistema mediterráneo. Autora: A. Espinosa.

X. Other Sources of Funding

None